

Negative pressure in shear thickening bands of a dilatant fluid

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We perform experiments and numerical simulations to investigate spatial distribution of pressure in a sheared dilatant fluid of the Taylor-Couette flow under a constant external shear stress. In a certain range of shear stress, the flow undergoes the shear thickening oscillation around 20 Hz. We find that, during the oscillation, a localized thickened band rotates around the axis with the flow. Analysis of the data shows that a major part of the thickened band is under negative pressure, which indicates that the thickening is caused by Reynolds dilatancy; the dilatancy causes the negative pressure in interstitial fluid, which generates contact structure in the granular medium, then frictional resistance hinders rearrangement of the structure and solidifies the medium.

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Introduction. A fluid with suspended particles have an apparent viscosity different from that of the medium fluid. If the total volume of particles is much smaller than that of fluid, the suspension behaves as a Newtonian fluid with the viscosity given by well known Einstein's viscosity formula [1]. In the case of high volume fraction, the fluid exhibits non-Newtonian behaviors. The viscosity decreases with increasing shear rate (shear thinning)[2] in many materials such as mud, paint, and ketchup. Shear thickening behaviors, i.e., increasing viscosity with shear rate, are also observed in other suspensions. In the extreme case such as the dense suspension of starch particles in water, the fluid almost solidifies under shear stress; the viscosity increases discontinuously by orders of magnitude [3–5]. They are often called “dilatant fluid” due to apparent analogy to Reynolds dilatancy of granular media [6], and their fascinating and unintuitive behaviors are popular subjects for science demonstrations [7–11].

The mechanism of discontinuous shear thickening (DST) is still under debate. A promising explanation is related to the dilatancy and jamming[12]. As stated in the Reynolds principle of dilatancy, dense granular media must dilate when they deform. If a suspension is confined, the dilation leads to jamming and then shear stress abruptly increases. In experiments performed with parallel plate or cone-plate geometry on a stress-controlled rheometer, suspensions dilates at the onset of shear thickening: the sample exerts an positive normal force between the plates [13]. The shear thickening has been also found to disappear if the normal force is removed from the boundary by controlling the gap between plates [4, 14].

It is numerically [15] and experimentally [?] demonstrated that contact friction between particles is important for DST in a shear flow of dry granular systems. The frictional contacts is found to be essential for DST also for the hard-sphere suspensions[16–18]. With these results, it is argued that DST is a consequence of jamming caused by dilatancy in a medium of frictional particles.

Lin *et al.* recently found that even in continuous shear thickening the contact forces between particles dominate hydrodynamic interactions[19].

In this Letter, we investigate the Taylor-Couette flow of a dilatant fluid by experiments and numerical simulations. We measure the off-center force on the axis and the local pressure at the wall of the outer cylinder. Our results indicate that the thickening is localized and that the negative pressure is dominant in the thickening region. We also perform numerical simulations for a three dimensional system using a fluid dynamics model developed by the authors [20, 21], which reveal the spatial distribution of pressure and viscosity in the flow. The results are consistent with the experiments. We find that there are two types of thickening bands, namely, the bands with positive pressure and those with negative pressure; the negative pressure band is dominant over the positive one, thus the suspension exerts attractive net force between the inner and the outer walls. The contact force network in the granular media is expected to span in the compressing direction. Contrary to this intuitive picture, we show that dominant thickening bands are under negative pressure and extend in the stretching direction, namely, the system jams due to tensile stress. Our local pressure measurement uncovers that the negative pressure is limited by the Laplace pressure, suggesting that the jamming under the tensile stress is sustained by the interstitial fluid.

Experiment. Our experimental setup is shown in Fig.1(a). The container consists of an acrylic outer cylinder (15 cm inner diameter) and an acrylic center rod (5 cm diameter) with an aluminum base plate and a lid; the gap h between the outer cylinder and the central rod is $h = 5$ cm. The center rod is supported with a couple of ball bearings; one is embedded in the base plate and the other is in the lid of the container. Constant torque in the counterclockwise(CCW) direction is applied on the center rod by a pair of weights on the opposite sides through steel wires wound around the rod. The weights are hung

through spring-dumper systems to reduce the vibration transmitted from the rod. The both weights are the same so that no net force should be applied on the rod. We use the weights in the range of $0.20 \sim 4.00$ kg, which corresponds to the external shear stress $S_e = 0.08 \sim 1.68$ kPa at the rod surface. In order to enforce the no-slip condition, the surface is lined with water-proof sand paper.

The suspension exerts off-center force on the center rod when the thickening is inhomogeneous. To measure this force, the upper ball bearing is supported at four points by load cells (Kyowa LMB-A-100N) as shown in Fig 1(b). We label them as “north”, “south”, “east” and “west” by their directions. Note that precise calibration in the off-center force measurements is difficult due to the friction at the contacts between a load cell and the ball bearing.

We also measure the normal stress σ_n at the surface of the outer cylinder by four pressure sensors (Kyowa PGM-G-02KG). They are located to the “north” of the center and aligned along the axial direction at intervals of 2 cm with the shallowest one located at 7.5 cm below the fluid surface; they are labelled as “ch1”, “ch2”, “ch3” and “ch4” from top to bottom.

We use the potatostarch particles (Hokuren) of irregular shape with their sizes distributed over the range of $5 \sim 30 \mu\text{m}$ [Fig.1 (c)]. The powder is dried for 24 hours at 60°C and 35% humidity. We prepare 39

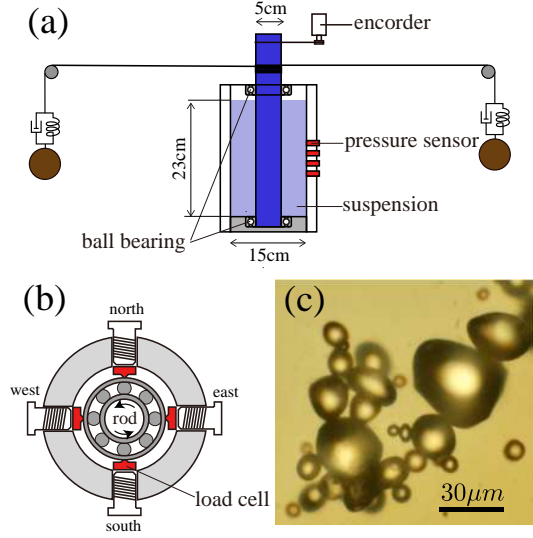


FIG. 1: (a) Schematic illustration of the experimental setup. A cylindrical container with a rotatable center rod is filled with starch-water mixture. A pair of weights are hung through steel wires wound around the center rod to give constant torque to the rod in CCW direction. The flow width h , i.e. the gap between the cylinder and the rod, is 5 cm. (b) Top view of the upper fixed end of the center rod. The ball bearing is supported at four points by load cells to measure the force acting on the rod. (c) Micrograph of the potato-starch particles.

wt% mixture with the density matched aqueous solution ($\rho = 1.76 \text{ g/cm}^3$) of cesium chloride (CsCl). The mixture fills the container up to $\ell = 23$ cm from the bottom with its surface open to the air.

For lower S_e , we observe steady rotation. However, when S_e exceeds a critical value around 0.1 kPa, the flow shows *shear thickening oscillation*, i.e. periodic alternation between the thickened and the relaxed states under a constant external shear stress[11]. The frequency is around 20 Hz and only weakly dependent on the external stress S_e and the flow width h .

Figure 2 shows the typical time developments of the off-center force f_{rod} on the center rod and the normal stress σ_n at the outer cylinder during the shear thickening oscillation. In Fig.2(a), the results from the four load cells at east, south, west, and north are shown for $S_e = 1.46$ kPa. Here, the time averages of the forces are subtracted from the measured data, and the positive value corresponds to compressive force on a load cell.

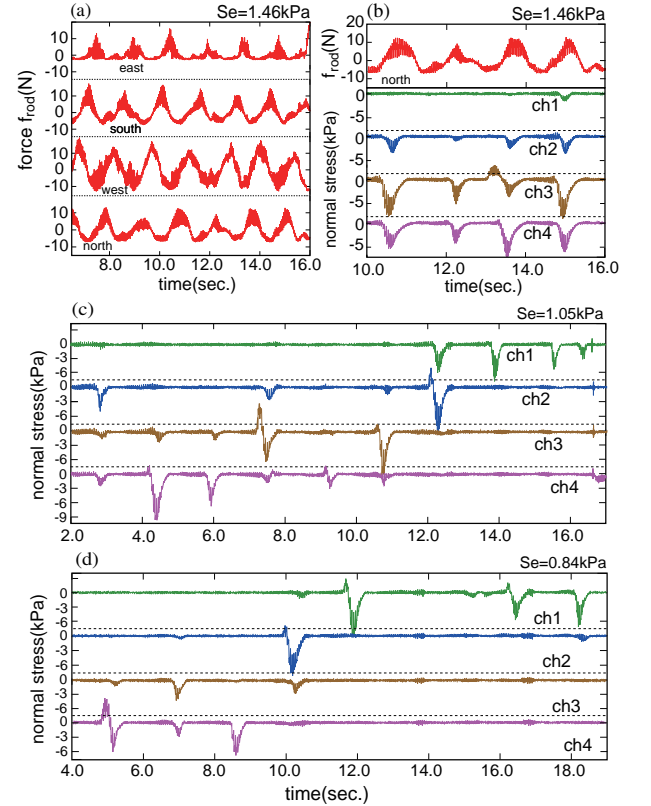


FIG. 2: Time evolution of the normal stress at the wall of the outer cylinder and the off-center force acts on the inner cylinder. (a) Off-center force for $S_e = 1.46$ kPa. (b) Off-center force and the normal stress for $S_e = 1.46$ kPa with the common time axis. (c,d) Normal stress for $S_e = 0.84$ kPa and for $S_e = 1.26$ kPa. The suspension is a 39 wt% mixture of potato starch and an aqueous solution of CsCl with a density of 1.75 g/cm^3 .

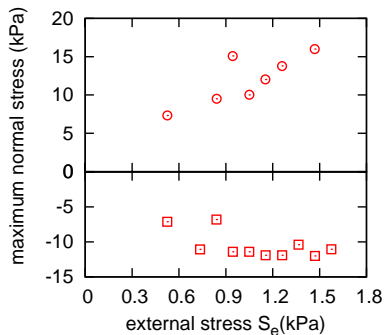


FIG. 3: Maximum peak values of pressure during the pulses as a function of external stress S_e . The plotted data are the largest peak values of positive (circles) and negative (squares) pulses observed by the sensors on the surface of outer cylinder during the shear thickening oscillation for each S_e .

The curves are roughly sinusoidal shape with the period $\tau_b \simeq 1.4$ s overlaid by the characteristic oscillation of shear thickening with the period $\tau_{sto} \simeq 0.05$ s. The time shifts between the plots for the neighboring load cells are $1/4$ of their periods, indicating that the direction of the off-center force rotates with the flow to the CCW direction.

Figure 2(b) shows the off-center force f_{rod} on the north load cell along with the temporal variations of the normal stresses σ_n measured by the four pressure sensors on the outer cylinder wall in the north. We observe periodic negative pulses in σ_n with the width $\tau_p \simeq 0.6$ s. They are slightly ahead of the peaks in the “north” component of f_{rod} . One may notice that the negative pulses are occasionally preceded by relatively weak positive pulses. It is also notable that many of the pulses are detected only by a couple of sensors, which reveals that the pressure fluctuation is localized in a region of a few centimeters thickness in the axial direction. These features are also seen in the σ_n data for $S_e = 1.05$ and 0.84 kPa in Fig.2(c,d).

Figure 3 shows the largest peak values of positive and negative pressure pulses, σ_{pos} and σ_{neg} respectively, observed by the sensors on the outer cylinder during the shear thickening oscillation for each value of S_e . It shows that σ_{pos} increases almost linearly with S_e . This result may corresponds to the linear correlation between the first normal stress difference N_1 and external shear stress reported by Lootens et. al [13], and in accords with the picture of the jamming mechanism. In contrast to σ_{pos} , σ_{neg} approaches the asymptotic value about -12 kPa after the initial decrease as S_e increases. The asymptotic value is close to the Laplace pressure $-2\gamma/R$, which gives -10 kPa for the average particle size of our potatostarch particles ($15 \mu\text{m}$) and the surface tension of water (73

mN/m).

Numerical Simulation. We performed three-dimensional (3- d) simulations for the Taylor-Couette flow using a fluid dynamic model of a dilatant fluid [20, 21]. The model is based on the incompressible Navier-Stokes equation with a phenomenological scalar field ϕ , which is supposed to represent the internal structure of the medium, such as the contact number of grains. The viscosity of the medium η is a function of ϕ , and local value of ϕ is driven by the shear deformation to the value ϕ^* determined by the local shear stress S of the medium. Assuming simple functional forms for $\eta(\phi)$ and $\phi^*(S)$, we have already demonstrated that the model reproduces basic properties of a dilatant fluids such as DST, the hysteresis upon changing shear rate, or instantaneous solidification by an external impact[20]. The shear thickening oscillation had been predicted by this model and was experimentally observed as had been predicted[21].

We employ the Highly Simplified Marker and Cell (HS-MAC) algorithm[22] for numerical simulations. As for the boundary conditions at the cylinder and the rod surfaces, we emulate the ones in our experiment, i.e. the no-slip fixed boundary at the outer cylinder and the no-slip boundary with a given average shear stress at the center rod, ignoring the mass of the rod. As for the boundaries in the rotating axis direction, however, we employ the periodic boundary condition for simplicity. The center rod rotates so as to give a constant shear stress S_e on the surface. We set diameter of the center rod $d = 2$, the flow width $h = 1.5$ and the fluid depth $\ell = 2.6$ in the dimensionless unit defined in [21]. With the parameters for the present medium, they are comparable with the ones for our experimental set up.

A uniform steady flow is unstable for the external stress S_e beyond a certain value. In Fig.4, we show snapshots of isosurface for $\eta = 2$ and for $p = \pm 1$ for $S_e = 1.5$, which is in the unstable regime. The results show that the viscosity and pressure distribution are not cylindrically symmetric. High viscosity regions tend to localize and finally form a single fan-shape thickening band, in which positive and negative pressure segments extend in different directions, i.e. the compressing and the stretching directions in the shear flow caused by the rotating center rod. As a result, the positive pressure segments always go ahead of the negative pressure segments at the surface of the outer cylinder. This is in agreement with the results of our normal stress measurement, where the positive pulses precede the negative pulses.

Our 3- d simulations show the shear thickening oscillation with a localized thickened band. The basic mechanism is the same with that in the 2- d simple shear flow[21]; in the S-shape shear rate-shear stress curve, there exists a range of the shear stress where the steady flow is unstable; under an external shear stress in the unstable range, no steady shear flow is possible and the

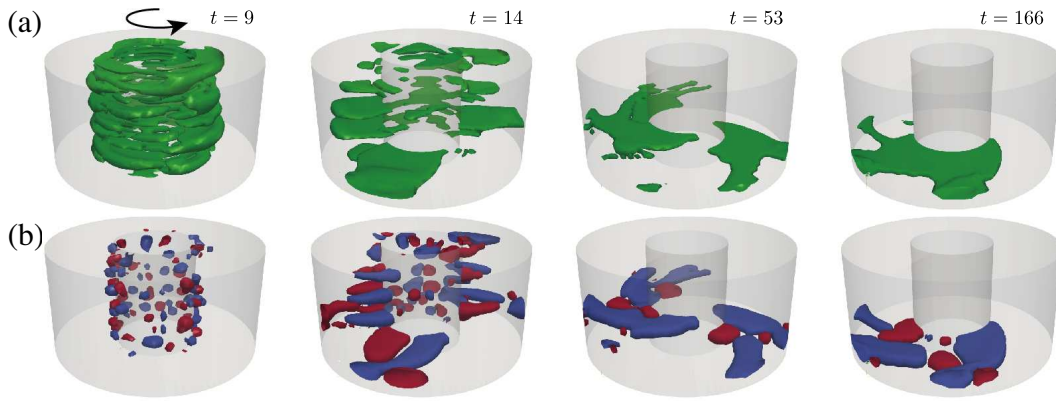


FIG. 4: Snapshots of the numerical simulation of Taylor-Couette flow for a phenomenological fluid dynamics model of a dilatant fluid [20, 21]. The depth of the flow is $\ell = 2.6$, the gap between the two cylinders is $h = 1.5$, and the external stress on the surface of center rod is $S_e = 1.5$. The arrow indicates the direction of rotation. (a) Isosurface of viscosity for $\eta = 2$. (b) Isosurface of pressure for $p = 1$ (red) and $p = -1$ (blue). The unit system is defined in [21].

system oscillates between the thickened and the relaxed states. In the shear stress thickening, the spatially uniform flow in the directions perpendicular to the shearing is intrinsically unstable because localized thickened bands can take most of the stress, leaving the rest of the medium in the unthickened state under low stress.

By the simulations of the Taylor-Couette flow, we can observe the dynamics of the thickened band; starting from the uniform relaxed state, the thickening regions first appear near the inner rod, where the shear rate is large. Then, some of the regions extend outwards, but eventually only one of them remains and reaches the outer cylinder. When the band reaches the outer cylinder, the flow decelerates. Then, it starts flowing again because the external stress is not large enough to keep the whole band in the thickened state. As the system starts flowing, the thickened band breaks in the outer regions, then the flow accelerates further until the shear stress causes thickening in the broken part of the band.

Discussions. The results of our numerical simulations and experiments are consistent and show that the thickening band distinctly has two types of segments: the positive pressure segment extending along the compressive direction of the shear flow and the negative pressure segment along the tensile direction. The remarkable finding is that the negative pressure segment is dominant in the shear thickening domain of the medium in the Taylor-Couette flow.

There are some differences between the two regions in the external shear stress dependence of the pressure as shown in Fig. 3; the maximum value of the positive pressure increases linearly with the external shear stress S_e , while the negative pressure does not seem to go below the Laplace pressure. Such linear dependence of the positive pressure has been reported also in Ref.[13], and may be understood in terms of the stress propagation along

force chains in the jammed granular medium. As for the negative pressure, it should be caused by the fluid in the interstitial space that tend to expand upon deformation of the medium due to Reynolds dilatancy. The fact that the negative pressure is limited by the Laplace pressure indicates that there exist the fluid-air interfaces possibly at the surface of micro-bubbles in the medium.

The shear thickening due to jamming in the negative pressure region has not been discussed, but it is natural for the granular medium with friction because the negative pressure in the interstitial fluid should increase contacts among the granular particles and the friction resists against rearrangement of the contact structure in the medium. It should be also noted that the shear thickening in the tensile deformation is easily observed in a simple demonstration by just pouring the starch-water mixture out of a cup, and the effect of shear thickening on drop formation in a granular suspension has been studied[23].

Although our experiments clearly show the existence of the negative pressure regions, it has not been reported in the literature; some experiments report only positive pressure when DST occurs[4, 13, 14, 24]. These experiments, however, do not observe the spatial variation of the pressure, but measure only total force on the upper plate of a cone-plate or plate-plate type rheometer, using small samples. In such measurement, the effect of the negative pressure may be hidden by the large positive pressure under strong external shear stress in the case where the value of the negative pressure is limited, even though the size of the negative pressure region is not small in comparison with that of the positive pressure.

In conclusion, our experiments and numerical simulations show that the negative pressure segment along the tensile direction is dominant in the shear thickening band of a dilatant fluid. The negative pressure in the

thickening bands indicates that the thickening is caused by Reynolds dilatancy; the negative pressure caused by the dilatancy generates contact structure in the granular medium, and the solidification of the medium is due to the frictional resistance against the rearrangement of the structure.

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